Multiple fluxoid transitions in mesoscopic superconducting rings

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The authors report magnetic measurements of fluxoid transitions in mesoscopic, superconducting aluminum rings. The transitions are induced by applying a flux to the ring so that the induced supercurrent approaches the critical current. In a temperature range near T_c, only a single fluxoid enters or leaves at a time, leading to a final state above the ground state. Upon lowering the temperature, several fluxoids enter or leave at once, and the final state approaches the ground state, which can be reached below approximately 0.5 T_c. A model based on the widely used time dependent Ginzburg-Landau theory for gapless superconductors can only explain the data if unphysical parameters are used. Heating and quasiparticle diffusion may be important for a quantitative understanding of this experiment, which could provide a model system for studying the nonlinear dynamics of superconductors far from equilibrium.

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The dynamics of phase slips (PS) in quasi-1D superconducting wires has long been studied intensively. Most observed phenomena have been explained with phenomenological models [1, 2] and the time-dependent Ginzburg-Landau theory (TDGL) for gapped superconductors [3], often with good quantitative agreement [4]. However, in those wires with diameter d of typically a few μm, the quasi-1D limit $\xi(T) \gtrsim d$ and $\lambda(T) \gg d$ was only accessible very close to the critical temperature T_c, where the coherence length ξ and penetration depth λ diverge. Due to various approximations, the microscopic validity of most of the theoretical models employed is also limited to the vicinity of T_c. Modern e-beam and nanotemplating techniques yield superconducting wires which remain quasi-1D down to $T = 0$, and can be thin enough ($d \sim 10$ nm) to study phase transitions due to quantum PS's. Such quantum phenomena, which have recently attracted tremendous interest [5], are possible when some action $S \sim \Delta E \Delta t$ is comparable to $\hbar$. Thus, the dynamical laws determining the time scale Δt are very important for understanding quantum effects. Furthermore, a better understanding of superconducting dynamics is interesting both for its own sake and for device applications, such as microbridge SQUIDs [6] or nanowire single photon detectors [7].

We have probed the dynamical evolution of the order parameter in mesoscopic Al rings at temperatures $T = 0.15 \ldots 0.9\ T_c$. Upon increasing the magnetic flux Φ_a threading a ring, the circulating supercurrent I increases quasistatically until it approaches the critical current, and the ring switches rapidly to a more stable state. We find that near T_c, the phase winding number n of the order parameter $\psi(x) = |\psi|e^{inx}$ changes by $\Delta n = 1$ at each transition, which can leave the ring in a metastable state well above the ground state. At lower T, Δn increases, and the final state approaches the ground state, while static states persist over a larger range of applied flux. Below $T \approx 0.5\ T_c$, the ring decays into the ground state over an increasing range of flux bias points. Such multiple fluxoid transitions were predicted based on the gapless TDGL equations [8] and observed at a single T [9]. Refs. [10] and [11] contain further related theoretical work on state selection and the effect of fluctuations on the dynamics of mesoscopic superconducting rings, albeit in a different parameter range. The TDGL model in [8, 9] can only reproduce our and previous ring data by choosing a numerical constant which is inconsistent with microscopic theory and transport measurements in the resistive state. The gapped TDGL theory may overcome this inconsistency. However in Al, both versions of TDGL are justified only extremely close to T_c, and we estimate that the neglected heating and quasiparticle (QP) diffusion may be important in our experiment. Lacking a tractable theory for superconducting dynamics far from T_c, it is common to use TDGL, sometimes well below T_c and in the gapless version. Thus, it is of considerable interest to understand to what extent those theories are adequate outside their theoretical range of validity, and to develop better theories. Experiments such as ours can be used for validation.

Unlike transport measurements on superconducting wires, where voltage reflects the mean phase winding rate, our experiment probes when phase unwinding ceases, once started. Thus, we explore fast dynamics without a high measurement bandwidth. Furthermore, DC transport measurements are prone to thermal runaway effects at low T, where the critical current is large and cooling is inefficient. In our case, heating is also significant, but localized in space and time due to the finite number of PS's. The absence of leads eliminates the need to consider proximity effects and boundary conditions at the contacts.

The results reported here are from 13 rings with radii $R = 1$ and 2 μm and line width $w = 70$, 115, 140 and

180 nm, measured in a single cool down. We measured I as a function of Φ_a and T by positioning a scanning SQUID microscope [12] over each ring individually. The samples were made by e-beam lithography using PMMA resist and liftoff. The 60 nm thick Al film was deposited using e-beam evaporation at a pressure of 10^{-7} mBar and a rate of 35 Å/s. Using $\xi_0 = 1.6$ μm for pure bulk Al [13] and $\xi(0) = 140$ to 180 nm depending on w, obtained as described below [see also Fig. 1(g)], we infer a mean free path of $l_e = 1.4\xi(0)^2/\xi_0 = 17$ to 28 nm. The measured T_c is 1.25 K.

Our SQUID sensor is designed as a susceptometer as discussed in [14, 15]. We positioned one of its pickup loops over a ring and recorded its response while sinusoidally varying Φ_a, generated by an on-chip field coil, at 0.6 Hz.To isolate the flux generated by I, we subtracted a background measured by retracting the SQUID from the sample. For each ring, we took 40 such Φ_a–I curves at each T, in steps of 10 mK — a total of more than 40,000 field sweeps containing 550,000 transitions.

We also collected data at a smaller Φ_a-amplitude, so that fluxoid transitions only occurred near T_c, to extract $\lambda^{-2}(T)$ and $\xi(T)$ [Fig 1(f, g)], as described in Ref [14]. Using fifth order rather than cubic polynomials to fit the low T Φ_a–I curves accounts for corrections to 1D GL at low T[16], and for imperfections in the rings, whose effect is biggest at large I. We also corrected for partial screening of the applied flux.

Fig. 1 shows a few individual field sweeps. Each branch of a Φ_a–I curve represents the response $I_n(\varphi \equiv \Phi_a/\Phi_0)$ of a state with fluxoid number n, with a linear response around $\varphi = n$, and a curvature caused by pair breaking at larger $\varphi - n$. $\Phi_0 = h/2e$ is the superconducting flux quantum. Since $w \ll R$ and $H_a \ll H_{c2}$, I_n and the transition points only depend on $\varphi - n$ to high accuracy. In the GL regime, $I_n(\varphi) = -(wd\Phi_0/2\pi R\mu_0\lambda^2)(\varphi - n)(1-(\xi^2/R^2)(\varphi-n)^2)$. At lower T, deviations from the cubic form become noticeable.

We have extracted the position φ_t of each transition and n for each branch, and computed $\phi_t(T) \equiv \langle|\varphi_t - n_i|\rangle$ by averaging over all transitions in each set of field sweeps. n_i and n_f are the n before and after each transition, as illustrated in Fig. 1(d). The ≤ 50 mΦ_0 rms-scatter of $|\varphi_t - n_i|$ around ϕ_t, which is partly due to sensor vibrations, is negligible for our purpose. The observed frequencies of $\Delta n \equiv |n_i - n_f|$ values yield a probability distribution as a function of T for each ring, as shown in Fig. 1(e). Since we find at most two consecutive integer values of Δn at any given T, the distribution is fully characterized by its mean $\langle\Delta n\rangle(T)$ [Fig. 2].

A GL stability analysis neglecting screening effects and fluctuations predicts $\phi_t = \sqrt{R^2/\xi_{GL}^2 + 1/2}/\sqrt{3} \approx R/\sqrt{3}\xi(T)$ [9]. In practice, ϕ_t as shown in Fig. 2 varies significantly between nominally identical rings, and is up to 30 % smaller than the measured value of $R/\sqrt{3}\xi(T)$, with the largest deviation for the smallest w. The variations suggests that imperfections contribute significantly to this discrepancy, but a deviation from GL theory far below T_c and thermal activation or tunneling may also play a role. The temperatures at which Δn changes also show significant scatter. However, they correlate strongly with the value of ϕ_t at that point for similar T. Consequently, the curves of $\phi_t(T) - \langle\Delta n\rangle(T) = \pm(\varphi_t - n_f)$ from all rings [Fig. 2] approximately collapse into a band whose width of ≈ 1 is largely due to the discreteness of $\langle\Delta n\rangle$. Below 0.5 T_c, $\phi_t - \langle\Delta n\rangle < 1/2$ can occur, which implies that the final state is the ground state with $|\varphi_t - n_f| < 1/2$. The lower T, the larger is the range of φ_t

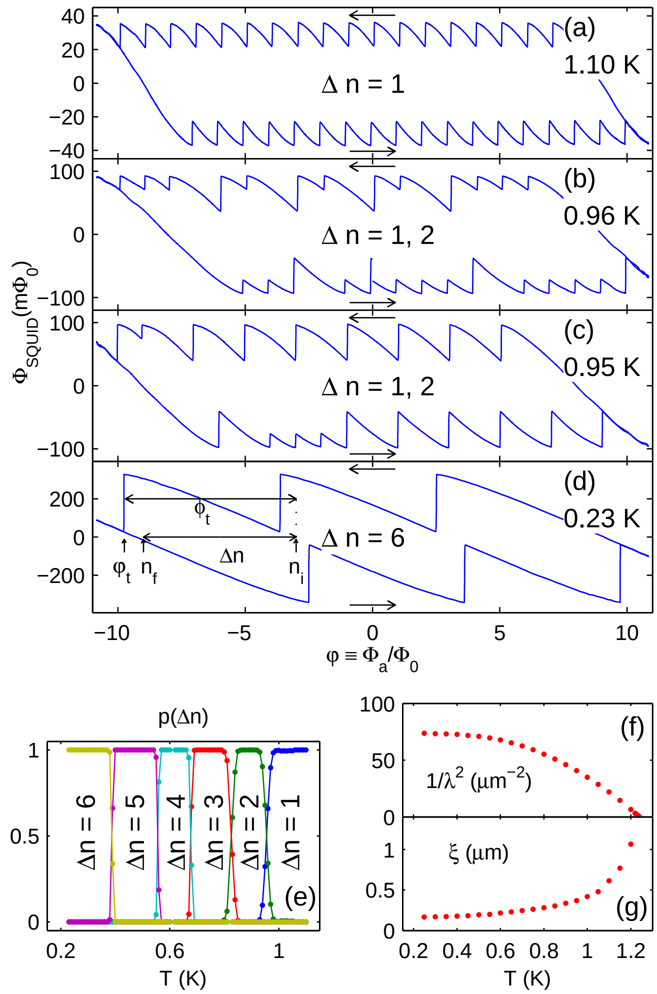

FIG. 1: (Color online) (a)-(d) Typical single-sweep Φ_a–I curves for a $R = 2\mu$m, $w = 180$ nm ring. The ring current I is proportional to the SQUID response Φ_{SQUID}, with mutual inductance $M_{coup} \approx 1.2$ Φ_0/mA for this ring size. At 1.1 K (a), $\Delta n = 1$ always. Around 0.96 K (b) and 0.95 K (c), $\Delta n = 2$ transitions become increasingly likely. At 0.28 K (d), $\Delta n = 6$, and the ring decays close to the ground state at each transition. The arrows indicate the field sweep direction and illustrate Δn, n_i, n_f and ϕ_t for the transition at $\varphi_t = -9.7$. (e) Probabilities for a change Δn in the fluxoid number. (f),(g) λ^{-2} and ξ as a function of T.

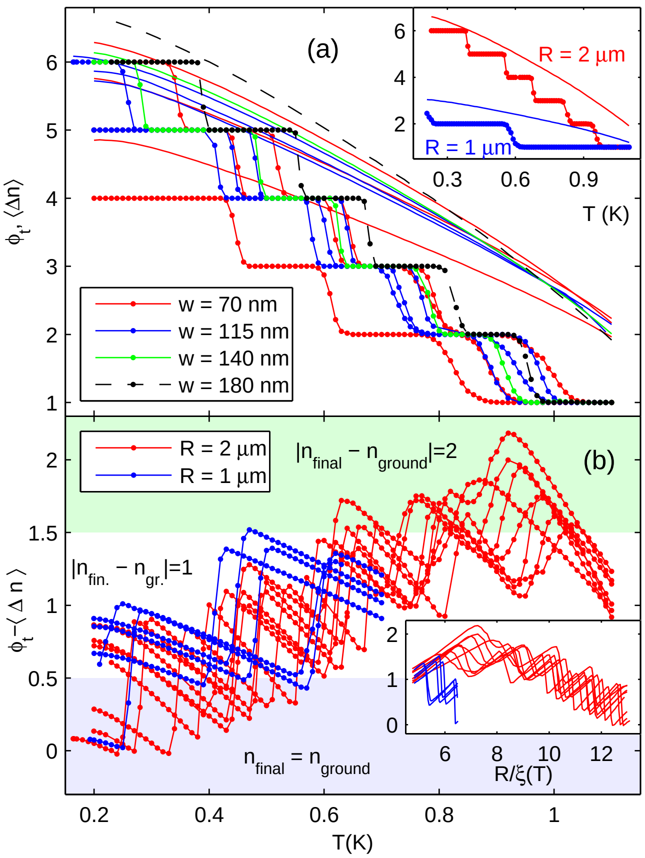

FIG. 2: (Color online) (a) Mean transition point ϕ_t (lines only) and $\langle \Delta n \rangle$ (lines with dots, each dot representing one dataset of 40 field sweeps). The inset shows the same data for just the ring from Fig. 1 and a $R = 1$ μm ring. (b) ϕ_t - $\langle \Delta n \rangle$ as a function of temperature for all 13 rings. The three shaded regions indicate the difference $|n_f - n_{ground}|$ between the n of the final and the ground state. Since $\phi_t - \langle \Delta n \rangle = \pm(\varphi_t - n_f)$, it encodes both $|n_f - n_{ground}|$, and the dependence of the final state on how far from an integer flux bias point a transition occurs. Comparison with the inset shows that $\phi_t - \langle \Delta n \rangle$ depends directly on the temperature rather than $R/\xi(T)$.

over which the ground state is reached. The absence of a clear w dependence of $\phi_t - \langle \Delta n \rangle$ vs. T indicates that the effect of the self inductance and the thermal activation barrier, both of which are proportional to wd, is small.

The results show no significant dependence on sweep frequency from 0.6 to 9.6 Hz. One concern is heating from the ~5 GHz, ~5 mΦ_0 Josephson oscillation applied to the rings by the SQUID. Varying the pickup loop–ring inductance M_{coup} by a factor 5 by retracting the SQUID about 2 μm had no significant effect. However, when the sensor chip touched the sample substrate, $\phi_t(T)$ flattened slightly below 0.35 K, changing by 0.1 at 0.2 K. Since this effect is negligible, the data was acquired with the SQUID touching the sample to avoid variations of M_{coup} due to scanner drift and vibrations.

We now turn to discussing our results in terms of the TDGL equations for gapped superconductors:

$$\begin{aligned} \frac{u}{\sqrt{1+\gamma^2|\psi|^2}} \left(\frac{\partial}{\partial t} + iV + \frac{\gamma^2}{2}\frac{\partial |\psi|^2}{\partial t} \right) \psi & \\ = (\nabla - i\mathbf{A})^2\psi + (1-|\psi|^2)\psi & \quad \text{(1a)} \\ \nabla^2 V = \nabla \cdot \mathrm{Re}(\psi^*(-i\nabla + \mathbf{A})\psi), & \quad \text{(1b)} \end{aligned}$$

where $\gamma = 2\tau_E \Delta_0(T)/\hbar$. $\Delta_0(T)$ is the equilibrium gap, V the electrostatic potential, and τ_E the inelastic electron-phonon scattering time. Without magnetic impurities, theory predicts $u = \pi^4/14\zeta(3) = 5.79$ [3]. Eqs. (1) are written in dimensionless variables, as defined in Ref. [17]. We only note that the unit of time is $\tau_{GL} = 2k_B T\hbar/\Delta_0^2$, and the dimensionless ring circumference is $2\pi R/\xi(T)$. The latter might lead to an indirect T-dependence. However, Fig. 2(b) shows only a weak R-dependence of the range of possible $\phi_t(T) - \langle \Delta n \rangle(T)$, whereas plotting $\phi_t - \langle \Delta n \rangle$ against $R/\xi(T)$ (inset) gives two distinct bands. Thus, the T-dependence of γ is more important than that of $R/\xi(T)$, to the extent that Eqs. (1) are applicable.

The mechanism underlying multiple PS's in mesoscopic rings was analyzed in Refs. [8, 9], based on numerical solutions of Eqs. 1 with $\gamma = 0$. Ref. [8] identifies two timescales: the duration of a single PS, τ_ϕ, and the relaxation time $\tau_{|\psi|}$ of $|\psi|$. If $\tau_\phi \lesssim \tau_{|\psi|}$, the phase may unwind by several 2π before $|\psi|$ can recover after the first PS. Further analysis for wires with finite γ [17] shows that τ_ϕ is related to the voltage drop $\Delta V = IR_{PS}$ across the PS center via the Josephson relation: $\tau_\phi = \Phi_0/\Delta V$, where R_{PS} is the resistance of the region around the PS over which the electric field decays and the QP current is converted to supercurrent. The extent of this region is given by the charge imbalance length $\Lambda_{Q^*} \propto \sqrt{\gamma/u}\xi(T)$. One obtains $\tau_\phi \propto \sqrt{u/\gamma}\tau_{GL}$ and $\tau_{|\psi|} \propto \gamma u \tau_{GL}$. The above expressions are valid for $\gamma \gg 1$. For $\gamma \leq 1$, one should replace γ with 1.

If treating u as a phenomenological adjustable parameter and setting $\gamma = 0$, one thus finds that $\tau_\phi/\tau_{|\psi|}$ decreases with increasing u, so that a larger u leads to larger Δn [8]. Reasonable agreement with a previous experiment at 0.4 K was obtained for $u = 48$ [9]. However, $u \gg 6$ is inconsistent with microscopic theory and experiments in the resistive state of quasi-1D superconducting wires [4, 18]. Those show that except very close to T_c, $\xi < \Lambda_{Q^*} \propto |T - T_c|^{-1/4}$. Some experiments even found a constant Λ_{Q^*}. When using Eqs. (1) with $\gamma = 0$, this implies $u < 1$ and that u decreases with decreasing T [8]. For example, Eqs. (1) with $u = 0.01$ and $\gamma = 0$ have been used to model the resistive state [18].

This inconsistency in the effective u arises because γ is negligible only in the gapless limit, where phonon (or magnetic impurity) induced pair breaking causes a fast reaction of the QP population to order parameter variations. For pure Al, this limit only applies for $T > T_c - 10^{-10}$ K. An increase of $\gamma \propto \sqrt{|T - T_c|}$ with

decreasing T on the other hand both reduces $\tau_\phi/\tau_{|\psi|}$ and increases $\Lambda_{Q^*}/\xi(T)$, in qualitative agreement with both ring and resistive-state experiments. However, Eqs. (1) neglect heating and QP diffusion, which is valid for $\tau_\phi, \tau_{|\psi|} \ll \tau_E$. Thus, even accounting for the slow charge imbalance relaxation by allowing $\gamma \neq 0$ only extends the theoretical validity to $T_c - T < 0.1$ mK for Al [19] [22].

The observation that $\varphi_t - n_f$ is nearly independent of R implies that the current after a transition is approximately inversely proportional to R. For wires carrying a quasi-dc bias current I on the other hand, phase slipping stops once I drops below some limit I_{c1}, at which $\tau_\phi = \Phi_0/R_{PS}I_{c1} \approx \tau_{|\psi|}$ [17]. Within the TDGL picture discussed above, this finite length effect could be explained if the normal-like length determining R_{PS} is set by circumference rather than Λ_{Q^*}, which is formally plausible since $2\Lambda_{Q^*} > 2\pi R$ at $T = 0$. However, QP's can only diffuse a distance $\sqrt{D\tau_\phi} \ll \pi R$ during a single PS, so that it seems questionable if the whole ring can contribute to R_{PS}. Alternatively, the local reduction of the critical current at the PS center could increase as more PS's occur, so that larger rings holding more fluxoids have a smaller final current. Well below T_c, where $\Delta_0 \gg k_B T$, the electronic heat capacity $c(T)$ is very small, while the kinetic energy density is large. Therefore, dissipating the latter in a PS will lead to significant heating, i.e. excitation of the energy mode of the QP population, which increases for larger R. Thermalization with the lattice occurs on the scale of $\tau_E \geq 50$ ns [19], whereas assuming that R_{PS} is at least that of a ring section of length ξ gives $\tau_\phi \lesssim 10$ ps. Thus, the electrons are approximately a closed system. If the kinetic energy was converted to homogeneous heating, the resulting electronic temperature would be given, very roughly, by max(T, 0.8 K), because $c(T)$ is small for $T \lessapprox 0.5T_c$. At this temperature, ϕ_t is still rather large, so that uniform heating would lead to smaller Δn than observed. However, given the short diffusion length $\sqrt{D\tau_\phi} \sim \xi \ll 2\pi R$, hot QP's will remain localized so that diffusive cooling may affect $\tau_{|\psi|}$.

A general description of time dependent superconductivity, similar to the Usadel equations for the static case, was derived in Ref. [19]. Unfortunately, the resulting system of equations for Δ, the Green's functions and electron distribution is rather complex. Approximate solutions for specific cases were studied in Ref. [20, 21]. Similar computations might allow a direct comparison between our data and microscopic theory. A much simpler first step could be a similar study as in [8] using Eqs. (1), but with γ rather than u as free parameter. Heating and QP diffusion could be accounted for through a local effective temperature governed by the heat equation.

In conclusion, we have shown experimentally that the final state after a field induced fluxoid transition in mesoscopic rings approaches the ground state for $T \to 0$. While localized heating likely is important, our results cannot be explained with uniform heating. Time dependent Ginzburg-Landau theory for gapped superconductors might give an adequate phenomenological model, however it is not microscopically justified in the experimental temperature range. Although our experiment is conceptually rather simple, a quantitative explanation is intriguingly complex and probably requires the incorporation of QP diffusion. Such a model may also provide insight in localized nonequilibrium effects on the superconducting dynamics in other cases, such as vortex motion or flux avalanches.

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